HOT CARRIER DYNAMICS IN THE X VALLEY
IN Si AND Ge MEASURED BY PUMP-IR-PROBE
ABSORPTION SPECTROSCOPY

W. B. Wang, M. A. Cavicchia, and R. R. Alfano

Institute for Ultrafast Spectroscopy and Lasers, and New York State Center
for Advanced Technology for Ultrafast Photonic Materials and Applications,
The City College of New York, Convent Avenue at 138th St, NY, NY 10031

Si is the semiconductor of choice for nanoelectronic roadmap into the next century for computer and other nanodevices. With growing interest in Si, Ge, and Si_mGe_n strained superlattices, knowledge of the carrier relaxation processes in these materials and structures has become increasingly important.[1] The limited time resolution for earlier studies[2,3] of carrier dynamics in Ge and Si, performed using N_d:glass lasers, was not sufficient to observe the fast cooling processes.

In this paper, we present a direct measurement of hot carrier dynamics in the satellite X valley in Si and Ge by time-resolved infrared(IR) absorption spectroscopy, and show the potential of our technique to identify whether the X valley is the lowest conduction valley in semiconductor materials and structures.

The details of the femtosecond visible-pump and IR-probe absorption set up has been reported elsewhere.[4] In this measurement, ~585 nm, ~400 fs pulses, obtained from the output of a synchronously pumped dye laser with a pulse-dye-amplifier, was used as a pump, and IR pulses tuned from 2.5 μm to 5.5 μm, generated in a $LiIO_3$ crystal by difference frequency method, was used to monitor the photoinduced IR absorption.

The measured change of the induced IR absorption at λ_{probe} =3.3 μm as a function of delay time between pump and probe pulses for intrinsic Si and Ge samples at room temperature are displayed in Figs.1 and 2, respectively. The salient features of the curves are the following: although the curve for Si is characterized by a long flat decay within the measured time range of ~30 ps, the curve for Ge has a short decay of ~6 ps followed by a long flat decay.

The measurements were also extended to other probe wavelengths of 2.8 μm, 4.5 μm and 5.2 μm for both Si and Ge. The time evolution of the induced IR absorption at those probe wavelengths was found to be similar as that for λ_{probe} =3.3 μm: a single flat decay appears with Si, and a short decay followed by a flat decay accompanies with Ge. It is clear that the different temporal behavior of the induced IR absorption for Si and Ge reflects their different carrier cooling processes and arises from their different band structures.

The induced total IR absorption is attributed to interconduction band absorption (ICA) by electrons from the lower X valley to the higher X valley separated by an energy difference in IR; free carrier absorption (FCA) by holes in the valence bands and by electrons in the conduction bands; and inter-valence band absorption (IVA) by electrons from the split-off to the heavy and light hole bands. Since the calculated values[3,5] of OD for IVA at the probe wavelengths are much smaller than the measured total OD, IVA can be neglected in our
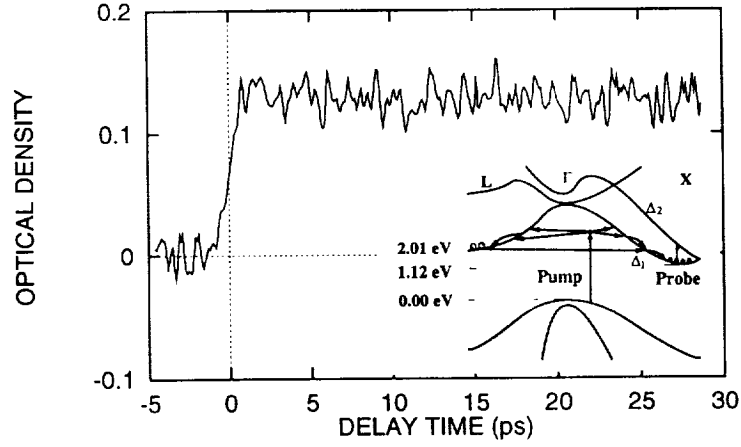
Figure 1. The measured change of the induced IR absorption as a function of delay time at $\lambda_{probe}=3.3$ μm in Si. The energy band structure of Si is shown in the inset.

consideration. The decay of FCA is flat because the recombination times of electrons and holes in Ge and Si are several order of magnitude longer than our experimental time scale. The long flat decay of FCA has been previously observed.[2-4]

The temporal characteristics of ICA in Si and Ge depends on their energy band structures. In Si, the lowest conduction band energy (1.12 eV measured from the top of the Γ valence band) is located at the X (Δ_1, Δ_2) valley[6] as shown in the inset of Fig.1. Hot carriers produced by 585 nm pump photons through an indirect transition can obtain sufficient energy to reach both the X and L valleys, from which they can scatter to each other as well as scatter inside each of the X and L valleys. Since the energy of the X valley minimum is lower than that of the L valley (2.01 eV)[7], after a few times of the X-L back-and-forward intervalley scattering, almost all of the electrons will finally scatter into the X valley. Those X valley electrons then decay to the bottom of the X valley through intravalley scattering, and stay there until they combine with holes in a few hundred nanoseconds.[8] Therefore, the induced ICA by X valley electrons from Δ_1 to Δ_2 (see the inset of Fig.1) has a long flat decay.

In contrast to Si, the lowest conduction band energy in Ge is located at the L valley[6] (0.66 eV) as shown in the inset of Fig.2. Electrons pumped by 585 nm photons through direct transition in the Γ valley obtain enough energy to undergo intervalley scattering from the Γ valley to the L and X valleys. The population of electrons in the X valley increases at first to its maximum through the Γ,L->X intervalley scattering. Since the minimum energy of the X valley (0.86 eV) is higher than that of the Γ and L valleys (0.80 eV and 0.66 eV, respectively), electrons stay in the X valley for only a short time, and then decay back to the Γ and L valleys. Therefore, the ICA in the X valley should have a short decay.

In order to quantitatively understand the change of the ICA with band structures, a parameter f_X, defined as the fraction of carrier density in the X valley relative to the total carrier density at the delay time corresponding to the flat absorption region, should be discussed. Since electrons in the conduction bands at the flat absorption region may be assumed to be in thermal equilibrium and characterized by a Boltzmann distribution, f_X can be written[9] as:

$$f_X = 1/[1+(t_X/t_L)(m_L/m_X)3/2 \exp(\Delta E_{XL}/K_BT)+(t_X/t_\Gamma)(m_\Gamma/m_X)3/2 \exp(\Delta E_{X\Gamma}/K_BT)] \quad (1)$$

where t_i and m_i are the lifetime and density-of-state effective mass of electrons in the ith ($i=\Gamma$, L and X) conduction valley, respectively; $\Delta E_{Xi} =(E_g)^X-(E_g)^i$ is energy difference between the X and ith vallys ($i=\Gamma$, L) at their minima; K_B is the Boltzmann constant and T is
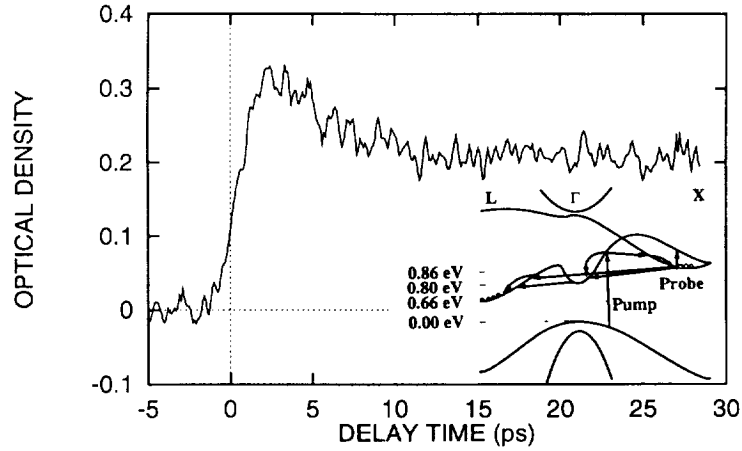
Figure 2. The measured change of the induced IR absorption as a function of delay time at $\lambda_{probe}=3.3$ μm in Ge. The energy band structure of Ge is shown in the inset.

the effective temperature of relaxed electrons (T=293 K). For Si, substituting ΔE_{XL}=-890 meV and other parameters[6,8] of t_X, t_L, m_X, and m_L into eqn.(1), f_X was calculated to be $[f_X]_{Si}$ ~1, which indicates that almost all of the electrons will decay and stay at the bottom of the X valley. Therefore, the ICA for Si is flat. For Ge, substituting values of ΔE_{XL}=200 meV and $\Delta E_{X\Gamma}$=60 meV, and other parameters[6,8] of t_i, m_i (i=L, Γ) into eqn.(1), f_X was calculated to be $[f_X]_{Ge}$ ~0, which means that the X valley holds almost no electrons at the long delay time. Since electrons stay in the X valley only for a short time, the ICA for Ge has a short decay.

The temporal behavior of the photoinduced IR absorption in a semiconductor can be used as a criteria to determine whether the X valley is the lowest conduction band valley. According to eqn.(1), materials with a conduction band energy minimum at the X valley ($\Delta E_{Xi}<0$ for i=Γ and L, therefore, f_X~1) should have a single flat decay, while materials with a conduction band valley minimum not at the X valley ($\Delta E_{Xi}>0$ for i=Γ or L, therefore, f_X~0) should have a short decay followed by a flat decay if the pump and probe wavelengths used can make the transition for ICA in the X valley. Over past several years, it has become a hot and controversial topic that certain Si_mGe_n strained superlattices may be converted from the indirect band gap of Si to a direct band gap via Brillouin zone folding.[1] The pump-IR-probe absorption technique presented here will be particularly useful for identifying this fundamental change of the lowest conduction band from the X valley to the Γ valley.

References

1. H.Presting, H.Kibbel, M.Jaros, R.M.Turton, U.Menczigar, G.Abstreiter, and H.G. Grinmeiss, Semicond. Sci. Technol., 7, 1127 (1992).
2. A.L.Smirl, J.Ryanlindle, and S.C.Moss, Phys. Rev., B18, 5489 (1978).
3. N.Ockman, R.Dorsinville, W.B.Wang and R.R.Alfano, IEEE QE-23, 2008 (1987).
4. W.B.Wang, K.Shum, R.R.Alfano, D.Szmyd, and A.J.Nozik, Phys. Rev. Lett., 68, 662 (1992).
5. R.Braustein, Phys. Rev., 130, 869 (1963).
6. O.Madelung, in "Semiconductors: Group IV and III-V Compounds, in Data in Science and Technology", edited by R. Poerschke (Springer, Berlin, 1991).
7. J.Weber and M.I.Alonso, Phys. Rev., B40, 5683 (1989-I).
8. B.R.Nag, in "Semiconductors probed by ultrafast laser spectroscopy", edited by R. R. Alfano (Academic, Orlando, 1984), Vol.I, Ch. 1.
9. W.B.Wang, R.R.Alfano, D.Szmyd, and A.J.Nozik, Phys. Rev., B46, 15828 (1992).